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13. ABSTRACT *(Maximum 200 words)*

The emphasis of the research in this program has been to investigate the use of UV lasers for surface diagnostics and for the probing of nanoscale surface physics. Our current program focuses on electron coupling and confinement on nanostructured surfaces, and the use of short-pulse nonlinear laser photoemission techniques to probe these phenomena. Specifically the program is to investigate low-dimensional, quantum confinement of electrons on surfaces patterned with angstrom-scale features. Electronic systems of reduced dimensionality are of interest for a variety of applications for future nano-electronic and -magnetic devices. For example, reduced dimensionality plays an important role in such important and timely phenomena as Q bits for quantum computing, and more generally in quantum-dot physics.

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THE INTERACTION OF SHORT ULTRAVIOLET-LASER PULSES WITH SURFACES: LASER PROBES OF NANOSCALE SURFACE

FINAL TECHNICAL REPORT

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Report of Inventions

A. Statement of the Problem Studied

The emphasis of the research in this program has been to investigate the use of UV lasers for surface diagnostics and for the probing of nanoscale surface physics. The program focuses on electron coupling and confinement on nanostructured surfaces, and the use of short-pulse nonlinear laser photoemission techniques to probe these phenomena. Specifically the program is to investigate low-dimensional, quantum confinement of electrons on surfaces patterned with angstrom-scale features. In addition, it aims to expand the applications and the technique of nonlinear surface photoelectron spectroscopy. Here the properties of the solid surface are sampled through the direction and kinetic energy of emitted electrons. Electronic systems of reduced dimensionality are of interest for a variety of applications for future nano-electronic and –magnetic devices. For example, reduced dimensionality plays an important role in such important and timely phenomena as Q bits for quantum computing, and more generally in quantum-dot physics.

1. Coherent Control of Electrons on Single-Crystal Metal Surfaces

Femtosecond laser pulses not only allow measurements of ultrashort lifetime events, but also provide an intense source of coherent radiation. Thus coherent control of electrons is possible with the ultrashort UV laser pulses. For example, coherent excitation of several eigenstates of the Rydberg series and quantum beating effect were observed on a Cu(001) surface by U. Höfer *et al.* A deeper understanding of coherence decay in metals is necessary for the application of coherent control of electron wave packets, which is important for the development of ultrafast optoelectronic devices and of direct relevance to spin electronics.

Time-resolved multiphoton photoemission spectroscopy has been proven to be a particularly powerful technique for probing carrier dynamics at surfaces and interfaces. Using

angle- and femtosecond time-resolved two-photon photoemission, we have measured the time evolution of the photoexcited electron population and polarization coherence at different states in the momentum space on Cu(111). Lifetime T_1 and pure dephasing time T_2 can be deduced from the spectrum linewidth and the cross-correlation traces, which were obtained by scanning the time delay between the probe and pump pulses. This is the first time that parallel-momentum-resolved lifetime has been conducted on a pure metal surface. The lifetime measured at the surface normal is 17 fsec ± 3 fsec. A secondary maximum in the lifetime at $k_{//} = \pm 0.08$ Å^{-1} was consistently observed and the reason behind this behavior is still under investigation. One hypothesis is that it is related with the coherent behavior of excited image electrons. At large angles, the lifetimes of image states drop very quickly, which can be explained by a combination of intra- and inter-band transitions. The inter-band transition was also confirmed with pump-beam-polarization dependence measurements.

By extending the similar time-resolved measurements to a stepped surface, one can get deeper insights on the interaction between electrons and surface nanostructures. This will be discussed in the next report.

2. A Coherent Model for Strongly Pumped Surface Electrons

As mentioned in the last section, intense femtosecond laser pulses make coherent control of electron dynamics possible. Also, coherent spectroscopic techniques that exploit the phase relationship between the excited electrons and the pumping beam gain more information than techniques that rely only on the measurement of intensities.

Recently, we have applied a three-level density matrix successfully for describing two-photon excitation of surface electrons, and used this model to fit the experimental data. In this model, the final state time response can be expressed as:

$$I(\Delta t)_{measured} = \int_{-\infty}^{\infty} \dot{\rho}_{ff}(t, \Delta t)dt \qquad (1)$$

where ρ_{ff} denotes the density matrix term for the final state and Δt is the time delay between the probe and pump pulses. Lifetime T_1 and dephasing time T_2 are used in this model as free parameters to fit the cross-correlation data. T_1 is responsible for the population dissipation and T_2 accounts for the decoherence process. Thus, for strongly excited surface electrons by a coherent field, lifetimes and dephasing times can be measured simultaneously.

For the special case of a continuum final state, such as photoemission, some simplifications can be made. Specifically, if the resolution of the energy analyzer (80 meV) is broad compared with the surface state linewidth, the detected signal includes not only resonantly ionized electrons from the intermediate image state, but also the electrons excited off-resonantly, which dephased faster with respect to the driving field than the resonantly excited electrons. Equation (2) is then modified to:

$$I(\Delta t)_{measured} = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \dot{\rho}_{ff}(t, \Delta t, h\nu_{probe}) \exp(-4\ln 2(h\nu_{probe} - h\nu_{probe}^{resonance})^2 / w^2) dt d(h\nu_{probe}) \qquad (2)$$

where w is the linewidth of the detector. For a relatively broad detector linewith, simulations show that the effect of pure dephasing time T_2 on the final signal is much smaller than the effect of the lifetime T_1 (only homogeneous broadening was considered in the simulations).

Coherent times can be controlled by ordered adsorbate overlayers or defects on the surface. In our measurements on stepped Cu(775) surface, step arrays can be viewed as line defects. Prior measurements have shown that the elastic and quasielastic scattering occurring at surface defects destroys optical phase coherence and effectively reduces the dephasing time; similar physics are expected to occur for step edges on our surface. Specifically, the pump pulse width in our experiment is ~30meV. Such a pulse will coherently excite a packet of image

electrons with a parallel momentum spread of $\Delta k_{//} \sim 0.087$ Å^{-1} around $k_{//} = 0$. The spatial extent Δd of such a packet will then spread in time as: $\Delta d = \sqrt{1/4\Delta k_{//}^2 + (\hbar \Delta k_{//} / m^*)^2 t^2}$. Thus a 14 Å terrace on Cu(775) will be traversed in less than 4 fsec. If electrons lose their phase coherence immediately after scattering by the step edge, the dephasing time will be very short even around $k_{//} = 0$; at larger $k_{//}$ the dephasing time will be even faster, making a simple rate-equation analysis appropriate. When $T_2 << T_1$, the density matrix analysis reduces to a rate-equation analysis,

$$\frac{dN(t)}{dt} = Ae^{-t/\tau} - \frac{N(t)}{T_1} \qquad (3)$$

where N(t) represents electron populations on the image state and T_1 is the image-state lifetime. The first term on the right hand side assumes a Gaussian pump pulse of width $(\ln 2) \cdot \tau$.

3. One Dimensional System: New Physics and Promising Fabrication Methods

The study of electronic and magnetic behavior of electrons under quantum confinement is important because of its fundamental implications and technical applications, as discussed earlier in this report. From a fundamental viewpoint, for example, Fermi-liquid theory based on a quasi-particle picture, breaks down for one-dimensional electron system because of the Peierls divergence in the electron-hole bubble. In this case, the Luttinger liquid model predicts several new phenomena, such as the vanishing of the Fermi edge, the appearance of charge and spin density waves, and the enhancement of electron-electron correlations. One-dimensional surface-type states have been observed on several systems, such as H/Ni(110) and Fe(100) c(2x2)Si surface alloy. In our work we expect that stepped surfaces will provide an ideal controllable system which may show one-dimensional effect since the two-dimensional symmetry is broken in the direction perpendicular to the step orientation. In addition, Himpsel *et al.* have proposed

that such effect might be adjustable by step decorations. The goal of our work is to find the effect of decoration on the one-dimensional state, which will finally lead to a controllable one-dimensional system.

We have recently shown for the first time that an occupied as well as an unoccupied one-dimensional surface state, as a result of step potential confinement and enhanced by step decoration, and the lifetime of such states can be measured. By a careful analysis of two-photon photoemission spectra, we have observed a one-dimensional occupied state on a stepped Cu(775) surface and located at ~270 meV below the Fermi level. An unoccupied local state also exists on the same surface; its lifetime is longer than the image state. Both states were measured to be dispersive along the step direction and non-dispersive perpendicular to the step orientation. Thus, steppped Cu(111) surfaces provide an potential system to study one-dimension electron behavior.

Our work has shown that very small amount of oxygen decoration on the stepped surface enhances the one-dimensional states. STM pictures by L. Bartels *et al.* indicate the oxygen is accumulated along the step direction, which is consistent with our local-state dispersion measurements. The origin of the states is attributed to the step-edge potential confinement. Furthermore, Anderson localiztion model was used to explain the effect of the disorder on the localized states. Upon oxygen exposure, the disorder effect was enhanced and the correlation between steps become weaker. Thus, the one-dimensional states are stronger upon oxygen exposure.

Further studies such as systematic time-resolved measurements on the newly observed states, STM observation of the clean and oxygen exposed Cu(775) surface, other decoration

measurements such as low temperature Xe deposition, polarization-dependence experiments may lead to new insights on the localized state.

Our current effort is focused on another quantum wire system: Ag decorated stepped Ni(755). Further studies will be on Ag decorated Pt(997) and In decorated stepped Si(111) surfaces. This will help to expand our knowledge of one-dimensional states and the physics of quantum wires, and finally, a deeper understanding of low-dimensionality physics, nanoscale micro-engineering and leading to novel electronic and magnetic devices.

C. List of All Participating Scientific Personnel (and those showing advanced degrees)

Hidong Kwak - Postdoctoral Research Scientist
Dan Mocuta - Postdoctoral Research Scientist
Gabor Nagy - Postdoctoral Research Scientist
Antonije Radojevic - graduate student
Shawn Shen - graduate student
Serban Smadici - graduate student

D. Publications & Presentations

1. X. J. Shen, H. Kwak, D. Mocuta, A. Radojevic, S. Smadici, and R. M. Osgood, Jr., "Observation of One-Dimensional States on Stepped Cu(775)," to be submitted to Phys. Rev.

2. X.L. Shen, H. Kwak, D. Mocuta, A.M. Radojevic, S. Samadici, "Observatin of a One-Dimensional State on Stepped Cu(775),"submitted to Phys.Rev. B..July 2000.

3. R.U. Ahmad, G. Nagy, and R.M. Osgood Jr., "Electron Cyclotron Resonance Plasma Etching of GaSb and GaSb-based Alloys," Appl. Phys. Lett., 77, 8/14/00.

4. G. Nagy, R.U. Ahmad, M. Levy, and R.M.Osgood, Jr. "Chemically Assisted Ion Beam Etching of Submicron Features in GaSb," Appl. Phys. Lett. 72, 1350 (1998).

Presentations

1. Optical Society of America, ILS-XV: Interdisciplinary Laser Science Conference, Santa Clara Convention Center, Santa Clara, CA, Sept. 26 - Oct. 1, 1999, (invited talk) "Optical Manipulation of Surface Electrons: Chemical Reactions and Quantum Structures."

2. Brookhaven National Laboratory, Upton, Long Island, New York, November 5, 1999, (invited talk), "Optical Manipulation of Surface Electrons: Reactions and Nanostructures," Richard M. Osgood, Jr.

3. University of Sherbrooke, Montreal Canada, May 4, 1999, (invited talk) "Image State Electrons: A Model System for Electron Transfer at Surfaces," R.M. Osgood, Jr.

4. CLEO/QELS '99, Baltimore, MD, May 23- May 28 1999, "Observation of Non-dispersive States on Stepped Cu(111)," H. Kwak, X.J. Shen, A.M. Radojevic, and R.M. Osgood, Jr.

5. CLEO/QELS '99, Baltimore, MD, May 23 - May 28, 1999, "Momentum-Dependent Ultrafast Dynamics of Image States on Flat and Stepped Cu(111) Surfaces," X.J. Shen, H. Kwak, A.M. Radojevic, and R.M. Osgood, Jr.,